# Fermiology of $Pd(dmit)_2$-based Molecular Solids Under Pressure

D. Graf, T. M. Murphy, E. C. Palm, R. L. Stillwell, W. Coniglio, J-H. Park, S. W. Tozer (NHMFL);
R. Kato, H. B. Cui (Condensed Molecular Materials Laboratory RIKEN, Saitama, Japan)

## Introduction

The organic compounds containing $Pd(dmit)_2$ molecules are Mott insulators under ambient pressure. In addition, spin-1/2 dimers formed from the $Pd(dmit)_2$ molecules add spin frustration as they create a 2D triangular lattice. Previous transport measurements of $EtMe_3P[Pd(dmit)_2]_2$ under 2 kbar of pressure have shown the suppression of the insulating state, revealing superconductivity with $T_c$ ~ 5K. [1] Similarly, $\beta$-$Me_4N[Pd(dmit)_2]_2$ has a $T_c$ of 5K when more than 6 kbar of pressure is applied. [2]

## Experimental

Skin depth measurements were performed at the NHMFL-Tallahassee facility in a 31 tesla resistive magnet using a top loading cryostat. The ~ 500 μm diameter sample was wrapped with a coil of wire and placed in the gasket of a diamond anvil cell. An oil medium was used for quasi-hydrostatic pressure and calibrated using the fluorescence peak of a small chip of ruby. For both measured samples, but especially for $\beta$-$Me_4N[Pd(dmit)_2]_2$, the samples were very thin (~ 10 μm) giving a small filling factor between sample and coil making high resolution for the measurements critically important.

## Results and Discussion

A TDO circuit was coupled to the sample coil and produced a resonant frequency of 50 - 200 MHz. The raw signal was amplified, mixed and recorded as the field was swept. Changes in the sample resistance appear as shifts in the circuit frequency (**Fig. 1a**). After removing a background from the resulting circuit frequency (Fig. 1a, upper inset), a fast Fourier transform of the data reveals three distinct orbit frequencies (Fig. 1a, lower inset) near ~ 4600 T for $EtMe_3P[Pd(dmit)_2]_2$ at ~ 2.3 kbar. This is reminiscent of another $M(dmit)_2$ compound, $Et_2Me_2N[Ni(dmit)_2]_2$, where similar frequencies were found but pressure was not required. [3] With a pressure of ~ 6.5 kbar, a single oscillation frequency of 4850 T (**Fig. 1b**) was observed for $\beta$-$Me_4N[Pd(dmit)_2]_2$. The oscillation amplitude is ~ 150 Hz with a resolution better than 1 ppm compared with the raw frequency. These subtle changes could not be seen with conventional transport measurements.

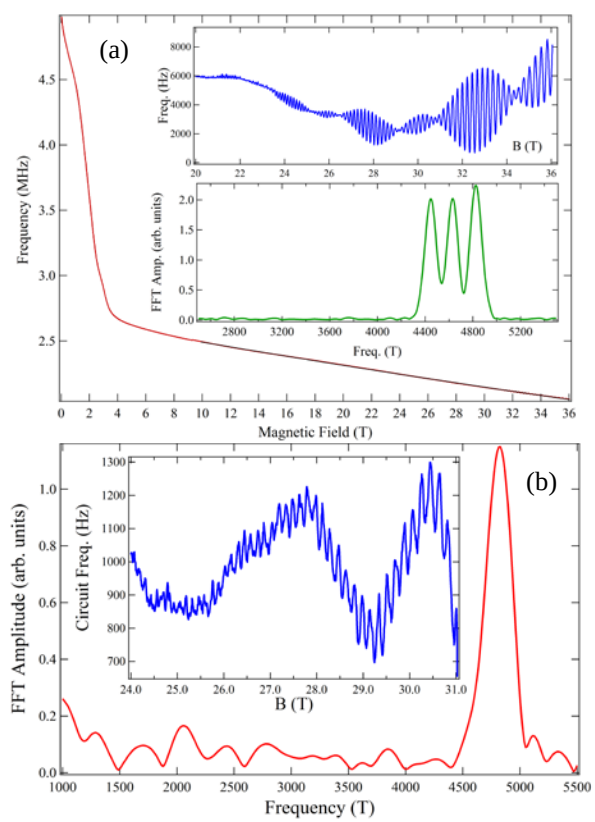


**Fig. 1**. Skin depth measurements of (a) $EtMe_3P[Pd(dmit)_2]_2$ and (b) $\beta$-$Me_4N[Pd(dmit)_2]_2$ under 2.3 and 6.5 kbar, respectively. The insets to (a) show the background subtracted circuit frequency shift and FFT. Inset to (b) shows the small oscillations produced in $\beta$-$Me_4N[Pd(dmit)_2]_2$.

## Conclusions

The FFT peaks of $EtMe_3P[Pd(dmit)_2]_2$ suggest magnetic breakdown between closely spaced surfaces. Despite the same dimer pairs of $Pd(dmit)_2$ molecules in $\beta$-$Me_4N[Pd(dmit)_2]_2$ we did not observe multiple oscillation frequencies suggesting differences in the Fermi surfaces at low temperatures.

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## References

[1] Shimizu, Y., *et al.*, Phys. Rev. Lett. **99** 256403 (2007).
[2] Kobayashi, A., *et al.*, Chem. Lett. **12** 2163 (1991).
[3] Tajima, H., *et al*, Solid State Commun. **88** 605 (1993).